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16. ABSTRACT

The O_3 measurements from the RAMS monitoring network in St. Louis in the upwind and downwind wind flow direction have been used to evaluate the impact of the urban plume of St. Louis. The time periods between April and October 1975 and 1976 have been utilized for this purpose. Incremental O_3 formation within the urban plume compared to regional O_3 background increases both in frequency and intensity from spring into midsummer and declines again in late summer and early fall. Episodic periods with substantial increments of O_3 formation in the plume over background occur especially during the summer months. Aircraft measurement of O_3 are used in combination with surface measurements to estimate the transport distance and width of the plume of St. Louis.

The significance is evaluated of incremental O_3 formation in the St. Louis plume on population exposures as well as on fumigations of field crops or forest in rural areas downwind. The strongest effects of O_3 formation in the plume occur in the prevailing wind direction. The results obtained from the St. Louis plume are discussed as they may apply to plumes from other cities especially in interior regions of the United States.

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SOME CHARACTERISTICS OF OZONE FORMATION IN THE URBAN PLUME OF ST. LOUIS, MO

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Abstract—The O_3 measurements from the Regional Air Monitoring System (RAMS) in St. Louis in the upwind and downwind flow directions have been used to evaluate the first day impact of the urban plume of St. Louis. The time periods between April and October 1975 and 1976 have been utilized for this purpose. Ozone formation in the urban plume has been evaluated with respect to the distribution of O_3 concentrations at RAMS stations downwind. The relationships between O_3 concentrations in the urban plume and such meteorological parameters as wind direction, wind speed, temperature, solar radiation intensity and maximum mixing heights have been investigated. Plume widths also have been estimated and related to plume O_3 intensities.

Incremental O_3 formation within the urban plume compared to regional O_3 background increases both in frequency and intensity from spring into midsummer and declines again in late summer and early fall. Episodic periods with substantial increments of O_3 formation in the plume over background occur especially during the summer months. Aircraft measurements of O_3 in the St. Louis plume are discussed by considering first day transport distances, plume widths and O_3 profiles beyond the outer stations in the RAMS network.

The significance of incremental O_3 formation is evaluated in the St. Louis plume on population exposures as well as on fumigations of field crops or forest in rural areas downwind. The strongest effects of O_3 formation in the plume occur in the prevailing wind direction. The O_3 in the plumes from all of the cities in an interior region of the U.S. is estimated to impact 5–10% of the rural lands in this region on any given day.

Key word index: Ozone, urban plume, effects of O_3 on rural areas.

INTRODUCTION

The experimental evidence for the formation of excess O_3 concentrations within urban plumes compared to O_3 concentrations in regional background air has been reviewed (Altshuller, 1986a; Spicer *et al.*, 1979). Incremental O_3 concentrations in urban plumes often have been observed in the 50–100 ppb range (Altshuller, 1986a) and even larger increments have been observed in large urban plumes (Spicer *et al.*, 1979).

Most of these investigations involved aircraft measurements of O_3 with traverses across urban plumes as they were transported downwind (Altshuller, 1986a). Such measurements are very useful in defining O_3 concentration profiles in urban plumes compared to the regional background O_3 concentrations. However the number of flight days available is usually very limited. Measurements of O_3 concentrations at relatively remote sites occasionally fumigated by urban plumes are also of interest (Altshuller, 1986a).

The O_3 measurements of the Regional Air Monitoring System (RAMS) at monitoring stations in and around St. Louis, MO obtained during the Regional Air Pollution Study (RAPS) provide an opportunity to investigate in more detail the characteristics of O_3 formation during the first day of transport within the urban plume of a reasonable isolated larger city in mid-continent.

Because of the incremental formation of O_3 in urban plumes above regional backgrounds, O_3 exposures are higher in downwind areas for people, agricultural crops and forest species. Therefore there is a need to quantitate the duration and frequency of elevated levels of O_3 in plumes over extended time periods. The results should have applications to several aspects of the health and welfare effects of O_3.

PROCEDURE

It is first necessary to evaluate how often an urban plume can be detected downwind of St. Louis. The daily distributions of the 1-h maximum O_3 concentrations during 1975 and 1976 at the Regional Air Monitoring System (RAMS) have been scanned. Twelve of the RAMS stations in the intermediate and outer rings are used for this purpose (Fig. 1). The daytime (0700–1359 h CST) vector average wind directions already available were used (Shreffler and Evans, 1982). These average wind vectors are used as an indication of the net drift of the mesoscale air mass during the daylight hours. It was decided that the maximum 1-h O_3 concentration at the selected downwind station should be at least 10–15 ppb higher than the mean concentration of the upwind stations in order to indicate the presence of a detectable plume downwind. This determination was based on the variability

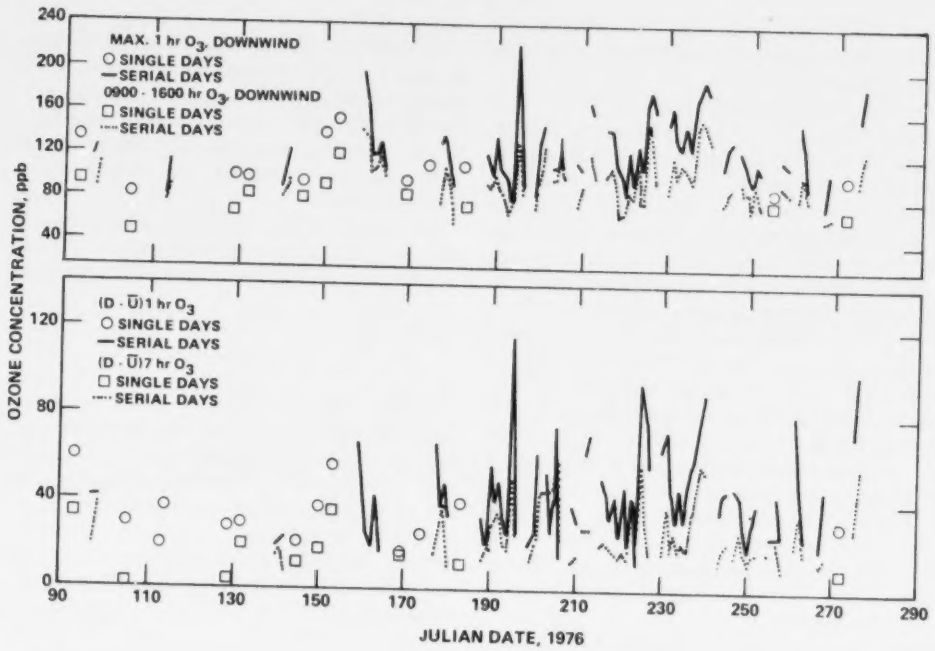


Fig. 3. Daily concentration values during 1976 of (a) max. 1-h O_3 downwind, (b) 0900–1600 O_3 downwind, (c) $(D - \bar{U})_{1h}$ and (d) $(D - \bar{U})_{7h}$ during April–October on days when urban plumes are detectable.

corresponding mean 0900–1600 h O_3 concentration downwind, (3) $(D - \bar{U})_{1h}$ and (4) $(D - \bar{U})_{7h}$ for the period April–October, are shown for 1975 in Figs 2 and 3 for 1976 for those days when plumes are detectable. Large day to day variations occur in the values of these quantities, in the values of $(D - \bar{U})_{1h}$ and $(D - \bar{U})_{7h}$. Clustering of the higher values into episodic periods is evident with respect to all of these quantities.

The maximum 1-h O_3 concentrations downwind, D_{1h}, exceeded 120 ppb on 41 of the 108 days in 1975 and on 44 of the 90 days in 1976 with detectable plumes. The means of the maximum 1 h concentrations upwind, U_{1h}, exceeded 120 ppb on only 1 day in 1975 and on 2 days in 1976 with detectable plumes. Although the mean values of U_{1h} did not exceed 120 ppb, the O_3 concentration did exceed 120 ppb at one or more upwind stations on 3 additional days in 1975 and 1976.

Examples of diurnal profiles at upwind and downwind stations are shown in Fig. 3 for 2 days with detectable plumes. The flat O_3 profiles during the late morning and the afternoon hours at the upwind stations are typical. On just over 40% of days with detectable plumes the maximum 1-h O_3 concentration occurs at midday, 1100–1300 h, as in the examples shown in Fig. 4. However, on 10% of the days with detectable plumes the maximum 1-h O_3 concentration occ[illegible]s as early as 1000 h or as late as 1700 h.

The monthly mean O_3 concentration increments of $(D - \bar{U})_{1h}$ daily and of $(D - \bar{U})_{7h}$ along with the frequency distributions of $(D - \bar{U})_{1h}$ values by month for April–October 1975 and 1976 are given in Table 1. The total number of days given, 189, is less than the total number of days with detectable plumes, 196, because of insufficient valid hours from 0900 to 1600 h to permit calculation of $(D - \bar{U})_{7h}$ on 7 days.

For $(D - \bar{U})_{1h}$ the two highest concentration values, 187 ppb and 161 ppb, occur during the latter part of summer of 1975. For $(D - \bar{U})_{7h}$ the two highest O_3 concentrations, 81 and 80 ppb, occur during the same time period. The 90th percentile O_3 concentration values for the periods April–October 1975 and 1976 of D_{1h} and $(D - \bar{U})_{1h}$ are 170 ppb and 80 ppb while the values for D_{7h} and $(D - \bar{U})_{7h}$ are 126 ppb and 45 ppb, respectively.

On a monthly average basis the ratios of $(D - \bar{U})_{7h}$ to $(D - \bar{U})_{1h}$ range from 0.42 to 0.68. From day to day the variations in this ratio are very large. These wide variations are associated with both the hour the maximum 1-h O_3 concentration occurs as well as the shape of the diurnal O_3 profile at the downwind station.

The number of days when urban plume widths, in km, are < 20, 20–29, 30–39 and ≥ 40 km are given in Table 2 with stratifications of these days among three ranges of $(D - \bar{U})_{1h}$ values. Broad plumes can occur in association with low intensity plumes, $(D - \bar{U})_{1h} < 40$ ppb. However broad plumes tend to occur with higher frequency in association with plumes having higher $(D - \bar{U})_{1h}$ values. As a result, plumes with

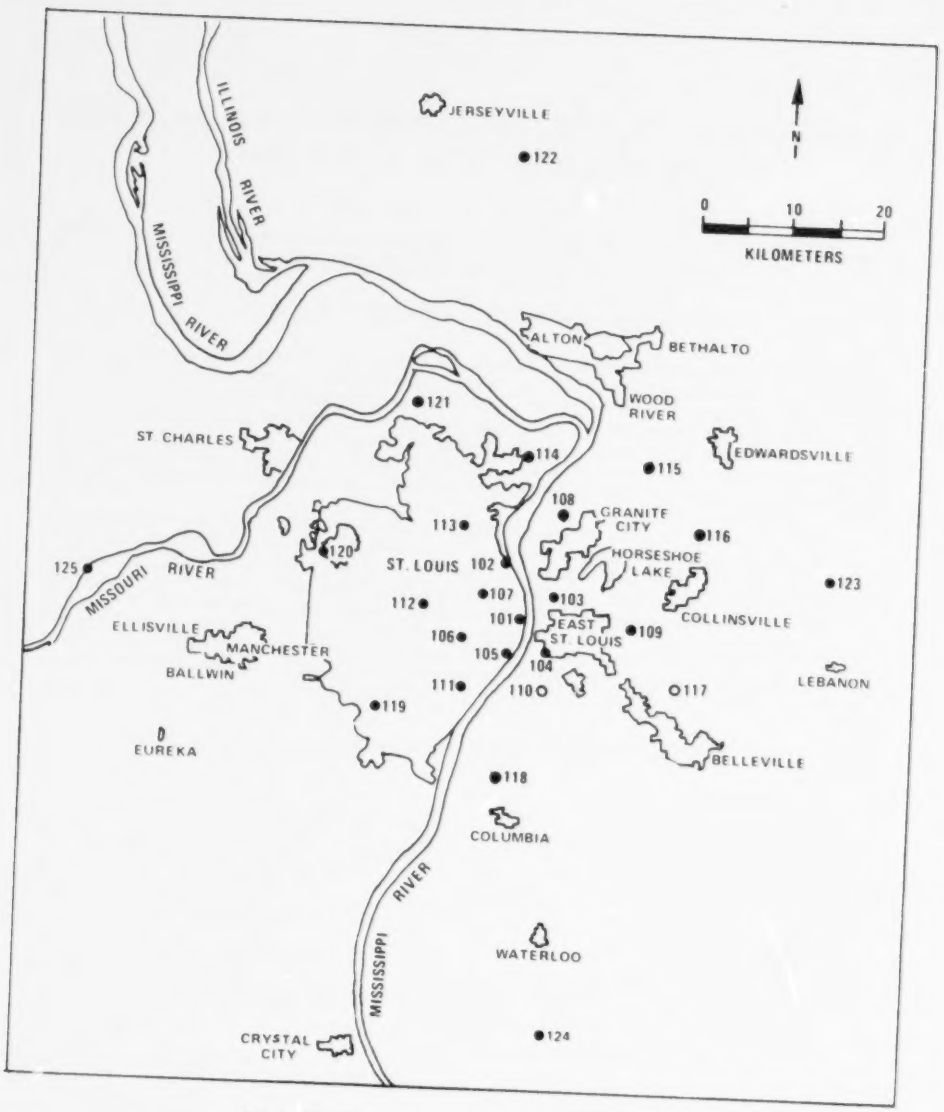


Fig. 1. The St. Louis region, rivers and RAMS sites.

of O_3 values among downwind stations and among upwind stations from day to day. It was also influenced by the size of the differences between O_3 concentration values shown to be statistically significant in a previous use of the same set of O_3 measurements (Altshuller, 1986b).

Based on meteorological considerations (as discussed later in the text), a negligible number of detectable plumes occur between November and March. Therefore this assessment is limited to days during April–October in 1975 and 1976.

A plume intensity parameter $(D - \bar{U})_{1h}$ is calculated for each of the days on which an urban plume is considered present. This quantity is defined as the difference between (a) the maximum 1-h O_3 concentration for the station nearest the center of the plume as it travels downwind into rural areas and (b) the mean of the maximum 1-h O_3 concentrations at upwind stations. The maximum 1-h O_3 concentrations at the outer or intermediate ring station downwind often selected are the highest values measured in the RAMS monitoring network. However, occasionally higher O_3 values were observed at inner ring stations, especially on those days when very low wind speeds occur.

Because of the use in agricultural crop yield studies of a 0900–1600 h period (Heck *et al.*, 1982, 1984) a plume intensity parameter $(D - \bar{U})_{7h}$ is also calculated. This quantity is defined as the difference between the mean of the 0900–1600 h O_3 concentrations for the station nearest the center of the plume traveling

downwind into rural areas and the average of the means for this time period of the O_3 concentrations at upwind stations.

The value of the concentration increment $(D - \bar{U})_{7h}$ was calculated from the O_3 measurements each day at the same downwind station at which the highest value of $(D - \bar{U})_{1h}$ was obtained. An alternative procedure would be to select the highest values for $(D - \bar{U})_{1h}$ and $(D - \bar{U})_{7h}$ separately each day. These two procedures have been compared using the group of days for which the $(D - \bar{U})_{1h}$ are in the 80th percentile and above. The mean value of the concentration increment $(D - \bar{U})_{7h}$ is 42 ppb by the procedure used. This value is 3 ppb less than the one obtained by the alternative procedure. However consistent use of $(D - \bar{U})_{1h}$ and $(D - \bar{U})_{7h}$ from the outer ring stations well into rural areas should give better representative rural values downwind. If the days involving highest values at outer ring stations are excluded the difference in the means for $(D - \bar{U})_{7h}$ from the procedures is 1.5 ppb.

From day to day the number of upwind stations with valid O_3 concentration measurements varies from one to nine stations with measurements usually available at three to five stations.

Plume widths immediately downwind of St. Louis have also been estimated. The minimum plume widths are estimated as the distances between the two downwind stations furthest apart but with D_{1h} concentrations 10–15 ppb above $\bar{U}_{1h}$ concentration values.

To relate the discussed plume intensity parameters to meteorological parameters, the 0700–1359 CST average temperatures, solar radiation intensities, wind directions and wind speeds are used (Shreffler and Evans, 1982) along with the daily maximum mixing height values (Schiermeier and Pooler, Personal communication).

Evaluations of the statistical significances of the differences in monthly mean values of $(D - \bar{U})_{1h}$ and of $(D - \bar{U})_{7h}$ are carried out by analysis of variance.

RESULTS

Ozone formation within the urban plume of St. Louis during April–October was detectable on 106 days (52% of total days in 1975) and on 90 days (43% of total days in 1976). Therefore, averaged over 1975 and 1976, O_3 formation within the urban plume is observable on just under 1 of 2 days during those months. The frequency of detectable O_3 in plumes increases during the late spring and summer months with detectable O_3 in plumes occurring on 9 out of 10 days during July 1975.

The values of the following quantities: (1) the maximum 1-h O_3 concentration in the central portion of plumes traveling downwind of St. Louis, (2) the

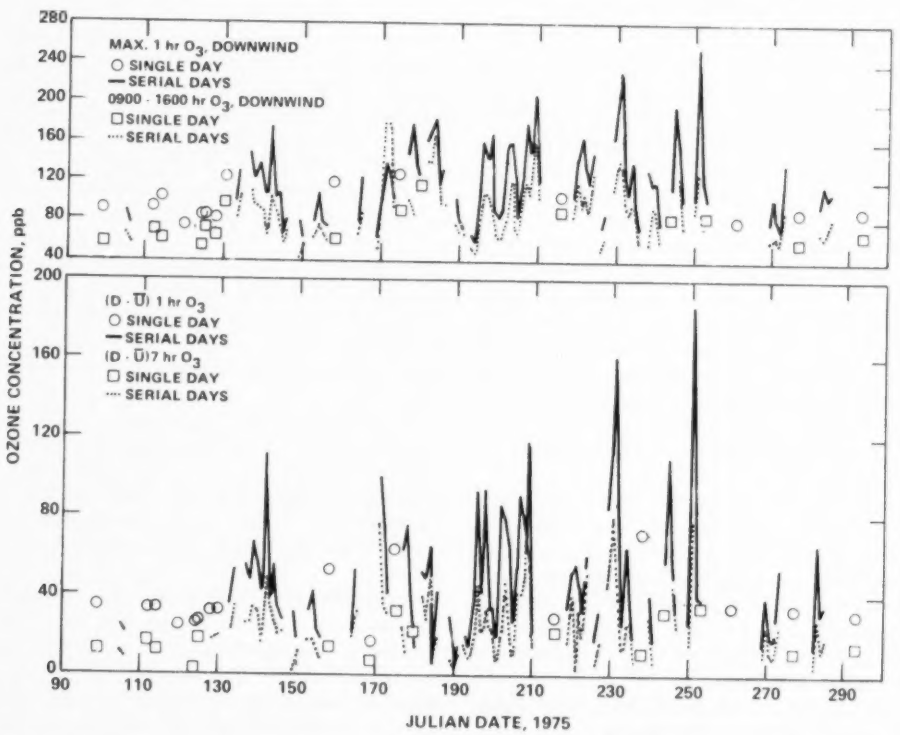
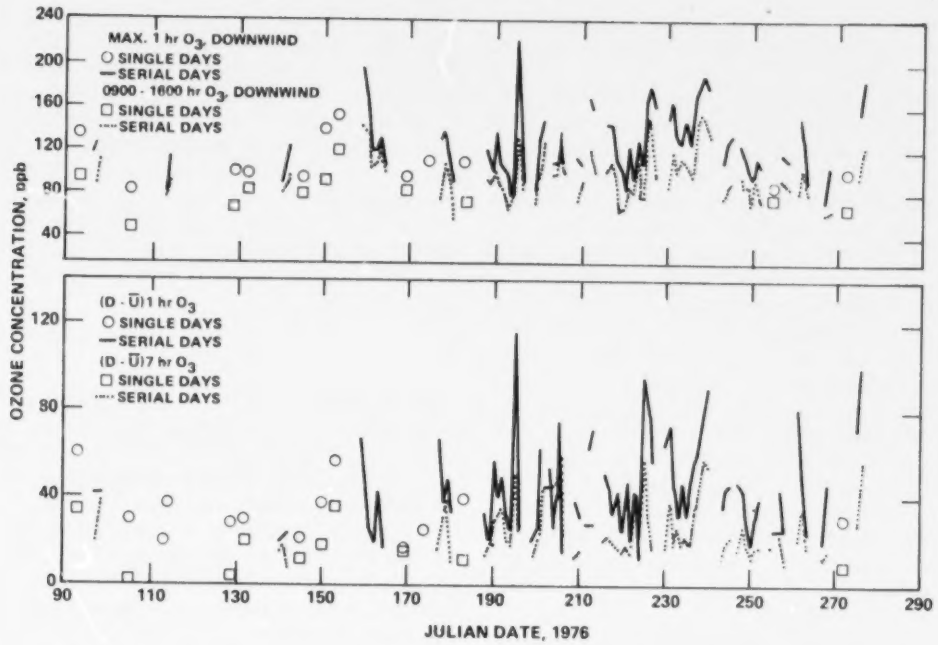


Fig. 2. Daily concentration values during 1975 of (a) max. 1-h O_3 downwind, (b) 0900–1600 O_3 downwind, (c) $(D - U)_{1h}$ and (d) $(D - U)_{7h}$ during April–October on days when urban plumes are detectable.



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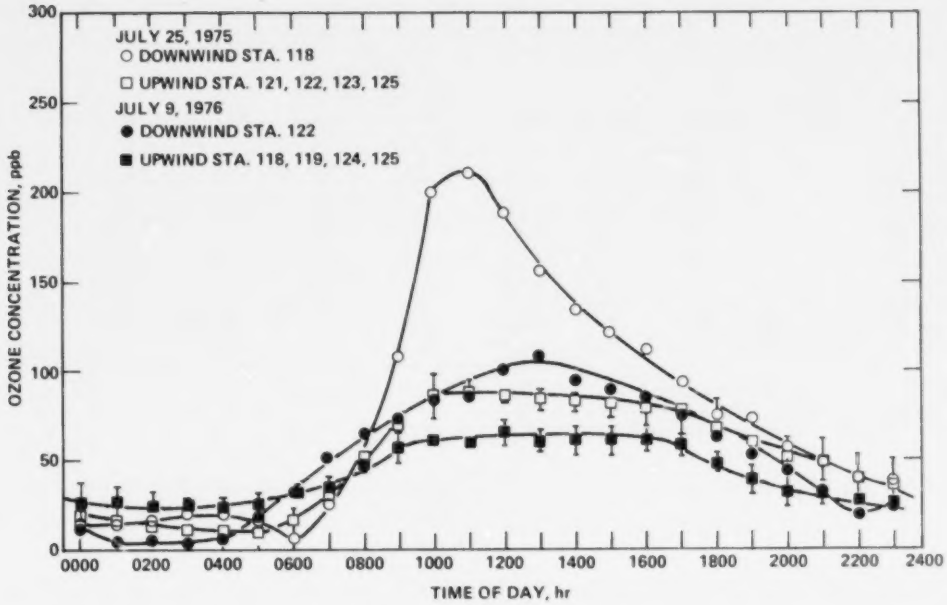

Fig. 4. Diurnal profiles of mean upwind O_3 concentrations and maximum downwind O_3 concentrations at RAMS stations on 2 days when urban plumes are detectable.

values of $(D-\bar{U})_{1h} \geq 80$ ppb almost always are broader plumes, widths ≥ 30 km, during 1975 and 1976.

The plume widths given in Table 2 tend to be underestimated because of (1) an insufficient number of monitoring stations and/or (2) invalid O_3 values at one or more of the downwind stations. Under even the more favorable conditions it is not possible to locate the edges of plumes. Upper limits of plume widths can be estimated immediately downwind based on the size of the urban, industrial areas of St. Louis. These upper limits vary somewhat depending on the direction of wind flow because of asymmetry in the dimensions of these areas. However the upper limit appears unlikely to exceed about 60 km.

DISCUSSION

The results demonstrate that the plume of a city such as St. Louis contributes substantial concentration increments of O_3 to rural areas downwind. Such contributions from urban plumes frequently occur on an episodic basis from spring to early fall. The more intense plumes, in terms of O_3 concentrations, tend to be broad and to occur during the summer months, especially July–August. The impact of O_3 in plumes on rural areas downwind of St. Louis tends to be strongest at or near the peak hour for O_3 formation downwind.

There may be an underestimation of the number of detectable plumes especially in early spring and fall months. Lower temperatures, lower solar radiation intensities and higher wind speeds in months such as April and October should result in slower rates of formation of O_3. If monitoring stations had been located at longer transport distances, enough reaction time may have been available to detect incremental O_3 in the plume.

Distribution of urban plumes as related to wind direction

It is of interest to determine to what extent the urban plumes may display directional asymmetry. An examination of the distributions of plumes with wind direction indicates that during April–October 1975 and 1976 windflows southeast to southwest tend to occur on a disproportionally large number of days. As a result plumes flowing through the northern intermediate and outer ring RAMS stations (114, 115, 121 and 122) occur on 57% of the days with detectable plumes. Much lower frequencies apply to flows from other directions especially for easterly or westerly flows.

Another approach to evaluating the influence of wind flow direction on O_3 has been utilized. A maximum 1-h O_3 concentration in excess of 120 ppb was exceeded on 85 days during 1975 and 1976 at one or more downwind stations on days when the urban plume was detectable. For this set of days the number of days on which maximum 1-h O_3 concentrations exceeded 120 ppb at several intermediate ring or outer ring stations was compared with the number of days such exceedances occurred at residential sites within St. Louis (Table 3). In addition the number of days with exceedances was compared for pairs of intermediate ring stations and for pairs of outer ring stations (Table

Table 1a. Frequency distributions and monthly means of $(D-\bar{U})_{1h}$ (first column) and $(D-\bar{U})_{7h}$ (second column) O_3 concentrations during April–October 1975 and 1976

Interval of $(D-\bar{U})O_3$ concentrations (ppm)	Month and year $(D-\bar{U})_{1h}$, $(D-\bar{U})_{7h}$ April 75	May 75	June 75	July 75	Aug. 75	Sept. 75	Oct. 75	Total
< 0.02	0,5	1,6	3,7	4,10	3,9	3,4	1,5	15,46
0.020–0.039	5,0	8,11	3,6	8,8	6,5	4,5	4,1	38,36
0.040–0.059	0,0	7,1	4,0	5,7	4,4	2,1	0,0	22,13
0.060–0.079	0,0	1,0	4,2	4,0	4,1	1,1	1,0	15,4
0.080–0.99	0,0	0,0	0,0	4,1	1,1	0,0	0,0	5,2
0.10–0.119	0,0	1,0	1,0	1,0	1,0	0,0	0,0	4,0
≥ 0.120	0,0	0,0	0,0	0,0	1,0	1,0	0,0	2,0
No. of values	5	18	15	26	20	11	6	101
Means, $(D-\bar{U})_{1h}$ O_3 (ppb)	28.4*	43.4*	46.6*	49.3*	53.0*	49.6*	36.8*	
Means, $(D-\bar{U})_{7h}$ O_3 (ppb)	12.8†	24.5*	23.7†	26.5†	27.6	26.6†	15.1‡	

* Differences in means statistically significant, $p \leq 0.001$.
† Differences in means statistically significant, $p \leq 0.01$.
‡ Differences in means statistically significant, $p \leq 0.05$.

Table 1b. Frequency distributions and monthly means of $(D-\bar{U})_{1h}$ and $(D-\bar{U})_{7h}$ O_3 concentrations during April–October 1975 and 1976

Interval of $(D-\bar{U})$ O_3 concentrations (ppm)	Month and year $(D-\bar{U})_{1h}$, $(D-\bar{U})_{7h}$ April 76	May 76	June 76	July 76	Aug. 76	Sept. 76	Oct. 76	Total
< 0.02	0,2	1,6	3,6	1,7	0,11	2,11	0,0	7,43
0.020–0.039	3,4	6,1	3,6	10,10	7,3	7,5	0,1	36,35
0.040–0.059	2,0	0,0	4,0	5,3	10,5	6,0	0,1	27,9
0.060–0.079	1,0	0,0	2,0	3,1	3,0	0,0	1,0	10,1
0.080–0.099	0,0	0,0	0,0	1,0	4,0	1,0	0,0	6,0
0.10–0.119	0,0	0,0	0,0	1,0	0,0	0,0	1,0	2,0
≥ 0.120	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0
No. of values	6	7	12	21	24	16	2	88
Means, $(D-\bar{U})_{1h}$ O_3 (ppb)	38.9†	26.4†	39.9†	44.4*	52.4*	38.0*	85.5	
Means, $(D-\bar{U})_{7h}$ O_3 (ppb)	21.8‡	13.4*	23.6‡	27.2*	29.2†	18.6*	42.0	

* Differences in means statistically significant, $p \leq 0.001$.
† Differences in means statistically significant, $p \leq 0.01$.
‡ Differences in means statistically significant, $p \leq 0.05$.

Table 2. Frequencies with which various urban plume widths occur in April–October 1975 and 1976 stratified by ranges in $(D-\bar{U})_{1h}$ values

Plume intensities $(D-\bar{U})_{1h}$, O_3 (ppb)	Year	No. of days having indicated plume widths (km) < 20	20–29	30–39	≥ 40	Total days
Less than 40	1975	37	6	8	4	55
	1976	19	7	8	9	43
40–79	1975	15	9	5	10	39
	1976	9	7	10	13	39
80 or greater	1975	0	2	6	4	12
	1976	0	0	1	7	8
Total days, 1975 and 1976		80	31	38	47	196

3). Because there were a number of days at each station when O_3 hourly measurements were incomplete or excess baseline drift occurred, the net number of paired days was well below 85 days.

The number of days with maximum 1-h O_3 concentrations in excess of 120 ppb are consistently larger at the northern downwind stations 114, 121 and 122 than at the other RAMS stations whether within the city or at intermediate or outer ring sites in other directions. At intermediate ring station 116 and at outer ring stations 123, 124 or 125, the number of days with maximum 1-h O_3 concentrations exceeding 120 ppb was smaller than at the stations in residential areas of St. Louis. Therefore, there is evidence for a higher exposure to O_3 at the northern edge of St. Louis and beyond than in other areas in and around St. Louis.

Table 3. Comparisons of number of plume days during 1975 and 1976 with maximum O_3 concentrations exceeding 120 ppb at selected pairs of RAMS station

Station A	Location of station	No. of days max. 1-h O_3 > 120 ppb	Station B	Location of station	No. of days max. 1-h O_3 > 120 ppb	Total no. of days with valid data
114	NW of city	17	120	NW area of city	12	34
114		22	112	NW center of city	13	53
114		21	106	near center of city	14	50
114		26	119	SW area of city	16	61
114		27	116	E of city	13	56
114		20	118	S of city	17	52
114		12	123	E of city	6	25
114		23	124	S of city	7	54
114		20	125	W of city	8	47
121	N of city	18	120		12	36
121		20	112		11	43
121		20	106		11	42
121		26	119		13	56
121		23	116		11	50
121		22	118		18	54
121		13	123		4	23
121		20	124		4	43
121		17	125		10	41
122	N of city	21	120		15	38
122	N of city	21	120		15	38
122		19	112		14	50
122		23	106		13	53
122		31	119		18	66
122		26	116		13	59
122		26	118		22	60
122		14	123		6	25
122		28	124		9	58
122		19	125		11	51
116	E of city	7	120		11	32
116		12	112		12	49
116		12	106		13	45
116		15	119		16	61
123	E of city	3	120		4	16
123		5	112		5	22
123		6	106		7	28
123		7	119		6	32
124	S of city	5	120		10	36
124		7	112		13	46
124		7	106		12	47
124		8	119		14	58
125	W of city	5	120		8	26
125		8	112		9	40
125		7	106		9	39
125		10	119		15	53

Relationship of O_3 plume intensities to meteorological parameters

The urban plume of St. Louis was not detectable on those days when the average daytime temperature, 0700–1359 h CST, was < 10°C. Urban plumes were detectable only on a few days with average daytime temperatures < 15°C. However, plumes were detectable on 90% of days with average daytime temperatures > 25°C and on all days when the average daytime temperature was > 30°C. On the 10% of days with average daytime temperatures > 25°C but with no detectable plumes the solar radiation intensities were low and/or the wind speeds high.

Additional comparisons have been made for those days with average daytime temperatures above 25°C and with solar radiation intensities ≥ 0.8 ly m^{-1}. The group of days with plume formation and $(D-\bar{U})_{1h}$ O_3 values < 40 ppb have been compared with the group of days with plume formation and $(D-\bar{U})_{1h}$ O_3 ≥ 80 ppb. With very little overlap the average daytime wind speeds on days with $(D-\bar{U}) < 40$ ppb prove to exceed 2.5 m s^{-1} while those on days with $(D-\bar{U})_{1h}$ ≥ 80 ppb are below 2.5 m s^{-1}. These meteorological conditions are consistent with sufficient transport time being available for air parcels to obtain sufficient inputs of precursors within St. Louis and to form substantial amounts of O_3 within the plumes moving past the downwind RAMS stations. It is possible if an additional arc of RAMS monitoring stations had existed further downwind, 80–120 km, that higher $(D-\bar{U})_{1h}$ values would have been observed at higher wind speeds because there would have been longer transport times available for O_3 formation.

When $(D - \bar{U})_{1h} \geq 80$ ppb the average daytime temperature always exceeds 20°C and exceeds 25°C on all but 2 days. The average daytime wind speed is $< 2.4\ m\ s^{-1}$ on all but 1 day. The wind flows are from the southeast to southwest on 63% of these days resulting in the plumes crossing the northern stations 114, 115, 121 and/or 122.

The mean values obtained from the average daytime temperatures, 27.5°C, and from the average daytime solar radiation intensities, 0.91 ly m^{-1}, for this group of days when $(D - \bar{U})_{1h} \geq 80$ ppb are only moderately higher than the corresponding values on all days. The mean of the maximum daily mixing heights for these days, 1700 m, is almost identical with the mean of the maximum mixing heights for all days. In contrast, the mean wind speed of 1.7 m s^{-1} for this group of days is 1–2 m s^{-1} slower than the monthly average wind speeds.

Combined aircraft and surface measurements of O_3 in plumes transport downwind of St. Louis

Light aircraft flights providing O_3 measurements during traverses of the St. Louis plume as it is transported downwind have been reported during 4 days in 1975 and 1976 (Gillani and Wilson, 1980; White *et al.*, 1976, 1977, 1983a, b). The O_3 measurements and related results are given in Table 4 for these 4 days. The total O_3 concentration measured within the center of the plume is given along with the difference between this value and the background O_3 concentration. For the surface measurements this difference is obtained from the quantity, $(D - \bar{U})_{1h}$, already discussed above. The values for $(D - \bar{U})_{1h}$ of 30–40 ppb on 3 of these days are less than the monthly mean values (Table 1) while the value on 28 July 1975 of 120 ppb is well into the 90th percentile range. For the aircraft measurements the difference is obtained by subtracting the O_3 concentration measured at the edge of the plume during a traverse from that in the center of the plume.

The values for the plume intensities and plume widths, 40–60 km, obtained from the aircraft measurements tend to be larger than those obtained from the surface measurements. Explanations have been offered in an earlier section for plume widths being broader than estimated from surface measurements. The values for the O_3 concentrations from the aircraft measurements might be expected to be higher because the traverses are taken further downwind of St. Louis than the RAMS network thereby allowing more reaction time for O_3 formation. Additionally some of the aircraft traverses may be obtained near the optimum transport distance for maximum O_3 formation.

There is an indication of peaking of the O_3 concentration at intermediate distances downwind from the traverses on 18 July 1975, 11 August 1975 and 9 July 1976. The times and distances at which the traverses were obtained indicate that elevated plume O_3 concentrations continue to be observed at least 125–150 km downwind within plumes 40–60 km wide. On 9 July 1976 the urban plume was tracked to about 280 km downwind of RAMS Station 122. The O_3 profile at this distance and a previous one at about 160 km downwind were associated with different plume parcels than the earlier profiles (Gillani and Wilson, 1980). The O_3 concentrations within the plume were still about 30 ppb above background in the 280 km downwind profile obtained at about 2300 h. Less detailed aircraft measurements of O_3 in the St. Louis plume are available on a few additional days (Hester *et al.*, 1977; Spicer *et al.*, 1982). These results appear to be generally consistent with those discussed here.

Measurements of O_3 at a rural site outside of the RAMS network

A range of air quality and meteorological measurements have been made beyond the northern outer ring RAMS station at a rural site outside the village of Glasgow, IL 104 km NNW of the Gateway Arch of St. Louis (Ellestad, 1980; Rasmussen *et al.*, 1977). Ozone measurements are available on 29 days from 16 July 1975 to 14 August 1975. The maximum 1-h O_3 concentration exceeded 80 ppb on 7 days. For the days 8–10 August 1975 the elevated O_3 concentrations are discussed in detail and, based on direction of wind flow as well as on non-methane hydrocarbon and CCl_3F measurements, are primarily attributed to the impact of the St. Louis plume (Rasmussen *et al.*, 1977). On 12 August 1975 the elevated O_3 concentrations are attributed to a regional scale stagnation episode upon which the impact of the St. Louis plume at Glasgow is superimposed (Rasmussen *et al.*, 1977). The 1-h O_3 concentrations during the afternoon and evening are given in Table 5. The influence of the St. Louis plume on elevated O_3 concentrations occurs as late as 2000 h.

Estimate of surface areas impacted by urban plumes

It is important to estimate the surface areas impacted by the first day transport of O_3 in urban plumes. Plume widths obtained from aircraft traverses range from 40 to 60 km—wider plumes than estimated for the same days from the surface O_3 measurements (Table 3). However, plume widths in excess of 40 km have been obtained from the surface O_3 measurements (Table 2). Reasons for possible underestimation of plume widths from surface O_3 measurements have been discussed in a previous section. In addition, some moderate plume widening as the plume travels downwind is not unreasonable as mixing occurs near the edges of plumes. Therefore, plume widths of 40–60 km should occur frequently. If plume widths of 40–60 km are combined with downwind transport distances of at least 125–150 km the resulting estimated surface areas impacted range from 5000 to 9000 km^2. Somewhat longer downwind transport distances can occur because of faster wind speeds aloft (Gillani and Wilson, 1980; White *et al.*, 1977). The total land area subject to fumigation by the urban plumes in a region can also be estimated. A count was made based on the 1980 U.S. census for the area west of the Appalachian Mountains and east of 105° W longitude. Within this area of about

Table 4. Comparisons of urban plume measurements obtained from aircraft and surface measurements downwind of St. Louis, MO

Date	Location downwind	Time (CST)	RAMS measurements, O_3 (ppb) Center of plume	RAMS measurements, O_3 (ppb) $(D-U)_{1h}$	RAMS measurements, O_3 (ppb) Plume width (km)	Aircraft measurements, O_3 (ppb) Center of plume	Aircraft measurements, O_3 (ppb) $(D-B)$*	Aircraft measurements, O_3 (ppb) Plume width (km)	Reference
18.07.75	at RAMS 114	1100–1200	85	30	35–40	NA†	NA†	NA†	This paper
	50 km NE of 114	1057–1120	NA†	NA†	NA†	180–190	70–80	50	White *et al.*,
	75 km NE of 114	1248–1310	NA†	NA†	NA†	130	40	50	1976, 1977, 1983a
	110 km NE of 114	1531–1556	NA†	NA†	NA†	130–140	40–50	50	
	145 km NE of 114	1704–1734	NA†	NA†	NA†	100	40	50	
28.07.75	at RAMS 118	1200–1300	210	120	$\geqslant 25$	NA†	NA†	NA†	This paper
	20 km S of 118	1351–1421	NA†	NA†	NA†	330	150	60	White *et al.*, 1977
11.08.75	at RAMS 115	1200–1300	115	30	40	NA†	NA†	NA†	This paper
	68 km NE of 115	1145–1210	NA†	NA†	NA†	180	100	60	White *et al.*,
	124 km NE of 115	1342–1408	NA†	NA†	NA†	140	60	60	1977, 1983b
9.07.76	at RAMS 122	1300–1400	105	40	40	NA†	NA†	NA†	This paper
	45 km NNE of 122	1444–1500	NA†	NA†	NA†	130–140	60–70	40–50	White *et al.*,
	125 km NNE of 122	1855–1920	NA†	NA†	NA†	110	40	50	1983a, b

* From aircraft measurements $(D-B)$ based on O_3 concentration in center of plume minus O_3 concentration at edges of plume.
† Not applicable

4×10^6 km^2 the following number of metropolitan areas by population category are found: $\geqslant 1$ million, 17; 500,000–1 million, 17; 250,000–500,000, 37; and 100,000–250,000, 66. Applying the results discussed above for St. Louis to the 17 largest metropolitan areas in this region leads to an estimated summed area of fumigation of 9×10^4–18×10^4 km^2 during days in spring and summer months.

For smaller metropolitan areas the available measurements of O_3 in urban plumes are very limited (Sexton, 1983; Spicer *et al.*, 1982). For Springfield, IL, a city with a population of almost 100,000, a plume with incremental O_3 values of 20–30 ppb has been measured 75 km downwind (Spicer *et al.*, 1982). The plume width was 25 km. The augmented plumes of several adjacent small midwestern cities with total populations adding up to somewhat > 500,000 have been observed (Sexton, 1983). On 2 days the incremental O_3 values in these plumes were 20–30 ppb, with plume widths of 30–50 km at distances of 75–80 km downwind.

These results for smaller cities indicate that plume widths do not decrease in proportion to population. This effect is consistent with lower population densities within smaller cities than larger cities thus leading to higher surface areas per capita for smaller cities.

Scaling these plume results for cities of various sizes, it is estimated that all of these metropolitan areas combined could fumigate on any given day a total surface area ranging from 5 to 10% of the 4×10^6 km^2 of the region under consideration. However, because of shifts in wind direction different areas will be fumigated from day to day, much more than 5–10% of the area around cities will be fumigated over longer time intervals.

Potential significance of results on exposures to O_3 of population groups

The results from this study indicate that the population groups in many areas more likely to be subject to higher exposures to maximum 1-h O_3 concentrations about 120 ppb will be downwind, especially downwind in the prevailing wind direction. Urban plumes with incremental O_3 formation within them are substantially more frequent in the prevailing wind flow direction. This prevailing wind flow was from the southwest to southeast during May to October 1975 and 1976. As a result, the number of days with exposures above 120 ppb at stations 114 and 121 near the northern fringe of St. Louis exceeded those at stations within the city by 50–100% during 1975 and 1976 (Table 3). Prevailing wind flow from the south is also observed from the longer term climatological results available between 1951 and 1960 (Baldwin, 1968).

Other cities to the west and southwest of St. Louis have an even stronger prevailing wind direction from the south to southeast during the warmer months of the year. Cities located to the east and northeast of St. Louis tend to have prevailing winds from south to southwest (Baldwin, 1968). These latter results are

Table 5. Selected ozone concentrations measured at Glasgow, IL*

Date	O_3 concentration (ppb) CST 1500 h	1600 h	1700 h	1800 h	1900 h	2000 h	2100 h
08.08.75	120	130	125	110	85	80	49
09.08.75	63	67	70	85	90	92	72
10.08.75	72	87	95	92	80	62	50
12.08.75	75	74	72	83	100+	76	56

* From Rasmussen *et al.*, 1977.

consistent with the O_3 measurements obtained downwind in the Detroit study discussed above (Kelly *et al.*, 1986). Similar prevailing wind flow towards the northeast accounts for the measurement studies on O_3 transport downwind of the New York–New Jersey area into Connecticut and beyond (Bufalini and Lonneman, 1977). Therefore, in these regions of the U.S., population groups living to the north and northeast of cities are subjected to the higher exposures to O_3 from urban plumes.

Potential significance of urban plumes on fumigations of field crop or forested areas by O_3

The urban plume effect on O_3 concentrations downwind was observable especially on rural areas within a 65° arc to the north of St. Louis including RAMS stations 114, 115, 121 and/or 122. The number of days when the plume intercepted this arc, the mean concentrations of $(D-\bar{U})_{1h}$ and $(D-\bar{U})_{7h}$ for the time periods the plume intercepted this arc, and the mean concentrations of $(D-\bar{U})_{1h}$ and $(D-\bar{U})_{7h}$ after being adjusted for days with no plumes or plumes falling outside of this arc during each time period are all given in Table 6. These calculations are made for each month between April and October 1975 and 1976 as well as for a number of shorter time periods during these months. The adjusted mean monthly values of $(D-\bar{U})_{1h}$ during the months of June–September range from 9 to 22 ppb while those for $(D-\bar{U})_{7h}$ range from 5 to 14 ppb. For a number of multi-month periods the adjusted means for $(D-\bar{U})_{7h}$ average near 10 ppb. The adjusted mean values of $(D-\bar{U})_{1h}$ and of $(D-\bar{U})_{7h}$ can be substantially higher during periods of about half a month and for shorter periods. For example, comparing the three periods, August 1975, 18 August–1 September 1975, and 18–24 August 1975, the adjusted mean value of $(D-\bar{U})_{1h}$ increases from 22 to 40 ppb up to 70 ppb while the corresponding value of $(D-\bar{U})_{7h}$ increases from 13 to 22 ppb up to 38 ppb. The 65° arc was partially overlapped by the urban plume on a number of additional days but these contributions are not included in the calculation made for Table 6. For arcs in other flow directions the urban plume effect on rural areas downwind is substantially smaller. In other flow directions the number of plume days falling within equal or somewhat wider areas is no more than one-third of the number of days falling within the northerly arc.

Even higher incremental O_3 concentrations within the urban plume occur for shorter periods than discussed above, 1–3 day periods (Figs 1, 2). Such high concentration, short-term fumigations of rural areas downwind occur not only within the northerly arc discussed above but occasionally in other flow directions.

The urban plume effect within the 65° arc downwind also must be adjusted for the probability that the urban plume is fumigating the rural areas within successive 65° sectors of rings. The plume widths are assumed to average nearly 40 km as the plume leaves the city and to gradually broaden to 60 km at 150 km downwind. The probability of fumigation of a given area within the sector within 50 km of the city is nearly 1, about 0.6 for the sector between 50 and 100 km downwind and about 0.4 for the sector between 100 and 150 km downwind.

The O_3 measurements during aircraft traverses of the St. Louis plume indicate that the incremental O_3 concentrations estimated from the RAMS O_3 measurements are probably underestimates of O_3 as the plume is transported downwind. This possibility of underestimation appears especially likely at intermediate distances, 50–100 km, where peaking in O_3 concentrations downwind are observed in the aircraft traverses (Table 4). Peak O_3 concentration values 2–3 times higher than those observed as the plume leaves the city are observed at intermediate distances. Such an increase in intensity of O_3 in the plume compensates for the lower probability of fumigation of rural areas in the second compared to the first sector downwind.

It is also necessary to consider the profiles of O_3 concentrations across a plume. The profiles observed from the traverses by aircraft are usually relatively flat. Based on the measurements of O_3 at three or four RAMS stations located downwind in the prevailing wind direction, 114, 115, 121 or 122, the average O_3 concentration across the entire plume was 0.8 of the average $(D-\bar{U})_{1h}$ in value.

Based on the St. Louis plume results, more intense fumigations by O_3 will be likely to occur for shorter diurnal periods and for certain monthly or episodic periods during the summer. Therefore evaluation of the potential of urban plumes to cause adverse effects on crop yields or forest growth is especially sensitive to time-averaging. For effects on yield of field crops more knowledge is essential in evaluating responses to lower

Table 6. Impact of O_3 formed in St. Louis urban plume on rural areas, within 65° arc north of St. Louis, MO

Time period	Year	No. of days plume within arc	Mean ozone concentration (ppb) plume days $(D-\bar{U})_{1h}$	$(D-\bar{U})_{7h}$	adj. for entire period $(D-\bar{U})_{1h}$	$(D-\bar{U})_{7h}$
April	1975	4	28*	14*	4	2
May	1975	14	45*	25*	21	12
June	1975	10	53†	28‡	18	10
July	1975	8	60*	28†	17	8
August	1975	13	53†	31†	22	13
September	1975	9	52‡	28†	17	9
October	1975	4	42*	18‡	6	3
April	1976	6	39*	22†	8	4
May	1976	2	28	17	2	1
June	1976	8	35‡	25‡	9	7
July	1976	10	49†	32*	16	10
August	1976	11	51†	31†	19	12
September	1976	8	35*	19*	10	5
October	1976	2	85	42	6	3
18–24 May	1975	7	59†	32†	59	32
20–27 June	1975	6	69*	34*	52	26
18–24 August	1975	7	70‡	38‡	70	38
21–27 August	1976	5	67*	51*	48	31
18–30 May	1975	11	48*	25†	40	21
18 August–1 September	1975	10	60†	32†	40	22

* Difference in means statistically significant, $p < 0.001$.
† Difference in means statistically significant, $p < 0.01$.
‡ Difference in means statistically significant, $p < 0.05$.

concentration, long-term exposures compared to higher concentration, short-term exposures (U.S. Environmental Protection Agency, 1986).

The St. Louis results are consistent with O_3 measurements in prevailing wind direction downwind of other cities in the eastern U.S. (Bufalini and Lonneman, 1977; Kelly *et al.*, 1986). In such regions field crop yield surveys should detect the larger reductions in crop yields and possibly adverse effects on forest growth caused by O_3 exposures in those rural areas downwind of the prevailing wind flow directions from such cities in the late spring and summer months.

On an episodic basis high exposures to O_3 can occur with winds in the prevailing direction but can also occur with transport of the urban plume into rural areas in other wind flow directions. The 20 days within the 90th percentile of $(D-\bar{U})_{1h}$ values distributed as follows: northerly flow, 12 days, southerly flow, 6 days; easterly flow, 2 days. The highest value of $(D-\bar{U})_{7h}$ of 81 ppb with an associated $(D-\bar{U})_{1h}$ of 118 ppb occurred when the wind flow was toward rural areas to the south of St. Louis. Such short-term high level O_3 exposures can be of considerable importance with respect to growth of forest species such as eastern white pine as well as field crops (U.S. Environmental Protection Agency, 1986).

CONCLUSIONS

Ozone formation in the St. Louis urban plume during the first day of transport downwind is detectable on about half the days during April–October 1975 and 1976. The frequency and intensity of O_3 formation in the urban plume increase from spring into the months of July and August and decline again in the late summer and fall. The intensity of O_3 formation in the plume as defined by the quantity, $(D-\bar{U})_{1h}$, and especially by the quantity, $(D-\bar{U})_{7h}$, varies substantially by a factor of as much as 5–10 from day to day even during the months of July and August. Plume widths are also highly variable although plumes with large $(D-\bar{U})_{1h}$ values also tend to be broad. In general, plume intensity (including the profile of O_3 concentrations across the plume, plume width and/or plume direction) can have unique aspects on each day.

Plumes with widths of 40–60 km—containing excesses of O_3 well above regional O_3 background—can be transported at least 100–150 km downwind of St. Louis. This is demonstrated from both aircraft measurement studies and O_3 measurements at a rural site well downwind of St. Louis in the prevailing wind direction.

The maximum 1-h O_3 concentrations observed at stations in plumes traveling downwind in the prevailing wind direction exceed 120 ppb on 50–100% more hours than at stations elsewhere within the St. Louis urban area. Such an excess of hours exceeding 120 ppb cannot be demonstrated to occur in downwind directions other than the prevailing wind direction.

A moderate excess in the O_3 concentrations fumigating rural areas in the prevailing wind direction north of St. Louis has been calculated to occur on a monthly average basis during the late spring and summer months. Much larger O_3 excesses in plumes over the regional background occurring during certain shorter time averages can cause substantial fumigations of

rural lands in the prevailing wind flow direction. On individual days substantially elevated O_3 concentrations over background occur in plumes traveling in other directions, especially to the south of St. Louis.

It is estimated that, on any given day when O_3 is formed in urban plumes, 5–10% of the rural land area within a large interior region of the U.S. could be impacted by such plumes. The location of this 5–10% of land area will vary with shifts in wind direction. Longer time-averaging evaluation of the impact of such plumes regionally requires quantitative results on the frequency of which plumes of varying intensities and widths are transported in the various wind directions around individual cities. Even in the prevailing wind direction, the importance of fumigations by O_3 in plumes is very sensitive to the time-averaging of significance to the effect being considered. The incremental impact of O_3 generated within plumes above regional background suspected of contributing to reduced yields of field crops or adverse effects on forest species emphasizes the need for surveys downwind of cities in the prevailing wind direction during the summer months.

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